

(19)



Europäisches Patentamt  
European Patent Office  
Office européen des brevets

(11) Publication number:

**0 277 728  
A2**

(12)

## EUROPEAN PATENT APPLICATION

(21) Application number: 88300477.2

(51) Int. Cl. 4: D21H 3/38

(22) Date of filing: 21.01.88

(30) Priority: 30.01.87 US 8981

(43) Date of publication of application:  
10.08.88 Bulletin 88/32

(84) Designated Contracting States:  
BE CH DE FR GB LI SE

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(54) Drainage and retention aids for newsprint furnishes.

(57) A method for improving drainage and the retention of inorganic fillers, cellulosic fines and/or pigments in the production of paper and paper-like products, especially newsprint, by the use of polymeric drainage/retention aids prepared from 75 to 99.9%, by weight, acrylamide and 25 to 0.1%, by weight, acryloyloxyethyl trimethyl ammonium chloride.

EP 0 277 728 A2

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**"IMPROVED DRAINAGE AND RETENTION AIDS FOR NEWSPRINT FURNISHES"****BACKGROUND OF THE INVENTION**

This invention relates to the use of acrylamide/acryloyloxyethyl trimethyl ammonium chloride polymers as drainage/retention aids in the manufacture of paper and paper-like products.

In papermaking, retention is generally defined as the retaining of inorganic fillers, such as clay and titanium dioxide, as well as cellulosic fines within the paper sheet during manufacture, as on a fourdrinier machine. The retention obtained on the wire screen is a function of different mechanisms such as filtration by mechanical entrainment, electrostatic attraction, and bridging between cellulosic fibers and fillers. Since both the cellulose and many common fillers are electronegative, they are mutually repellant and, in the absence of a retention aid, the only factor tending to enhance retention is mechanical entrainment.

Generally, retention aids are used because of the high cost of pigments, such as titanium dioxide. The use of retention aids significantly increases the amount of pigments incorporated into a wet web. Additionally, retention aids can appreciably reduce the suspended material in a paper machine white water effluent stream, which reduces pollution and loss of pigment.

Retention aids are commonly used in the manufacture of specialty papers, such as bond paper and "publication type" paper, including newsprint, where high loadings of pigment and/or filler are required. The function of retention aids is to bind a filler to cellulosic fibers without mechanically blocking the pores of the paper sheet being formed, thereby not adversely affecting the drainage properties of the wet paper web. Drainage, as used herein, refers to dewatering of paper or paper-like products being formed, as on a fourdrinier machine. The instant polymers improve both the retention and drainage properties of paper furnishes, and are therefore referred to as drainage/retention aids.

The prior art discloses the use of polyacrylamides as retention aids wherein about 3 to 35% of the amide groups are hydrolyzed to carboxylic acid groups. It is essential for alum to be present for such retention aids to work effectively, and their use is generally restricted to narrow pH ranges.

Amphoteric polymers have also been used as retention aids. For example, see U.S. Patent 3,639,208, which discloses the use of hydrolyzed acrylamide diallyl quaternary ammonium polymers as retention aids. U.S. Patent 3,926,718 discloses the use of N-vinylpyrrolidone/water soluble monomer block-type polymers as retention aids, wherein suitable water-soluble monomers include dimethyl diallyl ammonium chloride, 3-acrylamido-3-methylbutyl trimethyl ammonium chloride, methacryloyloxy 2-hydroxypropyl trimethyl ammonium chloride, methacryloyloxyethyl trimethyl ammonium chloride, methacryloyloxyethyl trimethyl ammonium methosulfate, 2-acrylamido-2-methylpropane sulfonic acid, acrylic acid and salts thereof, methacrylic acid, 3-acrylamido-3-methylbutyl dimethylamine, acrylamide, methacrylamide, diacetone acrylamide, hydroxymethylated diacetone acrylamide, dimethyl-1-(2-hydroxypropyl) amine methacrylamide, and sodium styrene sulfonate.

U.S. Patent 4,147,681 discloses the use of acrylamide homopolymer and copolymer emulsions as drainage/retention aids.

With respect to retention, newsprint furnishes present a difficult situation because of contaminants and carry-over from pulping operations. Because of these impurities, conventional drainage/retention aids become inactivated due to the demand of the contaminants for polymers. U.S. Patent 4,070,236 discloses the use of phenyl-naphtanol-sulfur resins in conjunction with polyalkylene oxides as newsprint retention aids.

**SUMMARY OF THE INVENTION**

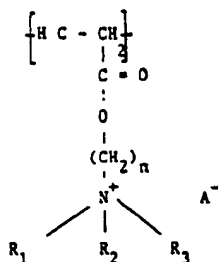
The instant invention provides an improvement in the art of papermaking in which the retention of cellulosic fines, fillers, pigments and other chemical additives is increased by the use of acrylamide (AM)-acryloyloxyethyl trimethyl ammonium chloride (AETAC) polymers. These polymers, which contain from about 75% to about 99.9% acrylamide and from about 0.1% to about 25% AETAC, by weight, and which also improve drainage properties, are especially effective in newsprint furnishes.



DETAILED DESCRIPTION OF THE INVENTION

This invention relates to a method for improving drainage and the retention of inorganic fillers, cellulosic fines, chemical additives and/or pigments in the production of paper comprising adding to an aqueous paper furnish an effective amount of a polymer comprising:

- (a) acrylamide or methacrylamide, preferably acrylamide; and
- (b) a monomer of the following structure:



wherein n is 2 or 3, preferably 2, wherein R<sub>1</sub>, R<sub>2</sub>, and R<sub>3</sub>, which may be the same or different, are C<sub>1</sub>-C<sub>8</sub> alkyl, preferably C<sub>1</sub>; and wherein A is Cl, Br, or CH<sub>3</sub> SO<sub>4</sub>, preferably Cl; wherein the weight percent of (a) is from about 75% to about 99.9%, preferably 85% to 99% and most preferably 90% to 98% and wherein the weight percent of (b) is about 0.1% to 25%, preferably 1% to 15% and most preferably 2% to 10%; and wherein said polymer has a reduced viscosity of greater than about 18 dl/g, preferably from about 18 to about 30 dl/g, more preferably 20-28 dl/g and most preferably 22-26 dl/g, measured at 0.5 weight percent using a 1.0 N NaCl solution at 30°C.

The instant invention additionally relates to the above described polymers, and to compositions comprising (a) a paper or paper-like furnish, preferably a newsprint furnish, and (b) at least one of the above described polymers.

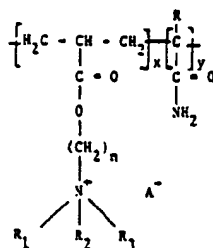
As used herein, the term "effective amount" refers to that amount of polymer which provides a drainage and/or retention improving quality to the furnish being treated. Generally, the polymer dosage, on an active basis, ranges from about 0.1 to about 10 pounds of polymer per dry ton of pulp.

While the instant polymers may be used as drainage/retention aids in any paper furnish, including furnish for fine paper, bond paper and publication-type paper, and in furnishes for the preparation of miscellaneous paper-like products such as liner board, they are especially effective in newsprint furnishes. As used herein, the term "newsprint furnishes" is defined as a furnish which is used to prepare newsprint paper, such as a furnish containing groundwood, mechanical pulp, Kraft pulp, Kraft sulfite pulp, semi-bleached Kraft pulp, thermal-mechanical pulp, deink stock and/or recycled paper.

The instant polymers may be added to a paper furnish at any convenient point to improve drainage and/or the retention of fines, fibers and additives on the paper web being treated. Generally, however, the drainage/retention aid will be added at a point immediately prior to the forming surface.

It is a particular feature of this invention that the introduction of the instant polymers into a papermaking furnish, particularly a newsprint furnish, will lead to improved drainage and/or improved retention of the cellulosic fines and various additives, including but not limited to pigment and fillers, over a wide range of pH and other chemical conditions. The desired drainage and retention improving characteristics of the instant polymers have been demonstrated in a wide variety of furnishes, as demonstrated by the examples included herein.

The polymers of the instant invention may be described by the following structure:



wherein R is H or CH<sub>3</sub>, R<sub>1</sub>, R<sub>2</sub> and R<sub>3</sub>, which may be the same or different, are C<sub>1</sub>-C<sub>8</sub> alkyl, n is 2 or 3, A is Cl, Br or CH<sub>3</sub>SO<sub>4</sub>, and x and y yield a polymer which is 75-99.9%, by weight, of the y moiety and 0.1-25%, by weight, of the x moiety. Preferably, R is hydrogen, R<sub>1</sub>, R<sub>2</sub> and R<sub>3</sub> are CH<sub>3</sub>, n is 2 and A is Cl. In this instance, the polymer is a copolymer of acrylamide and acryloyloxyethyl trimethyl ammonium chloride (AETAC), which is preferred polymer. Further, the inventor has found that at least 75%, by weight, of the polymer must comprise the acrylamide-like y moiety, preferably about 85% to about 99%, and most preferably about 90% to almost 98%. The inventor has also found that molecular weight, as determined by reduced viscosity, is critical. The instant polymers must have a reduced viscosity greater than 18 dl/g and preferably from about 18 to about 30 dl/g, more preferably 20-28 dl/g and most preferably 22-26 dl/g (0.5% in 1 N NaCl at 30°C).

Generally, about 0.5 pounds of the instant polymer (active basis) per ton of dry pulp will be used. The instant drainage/retention aids will seldom be used in amounts of less than 0.1 pound per ton of dry pulp, and in normal practice, for economic reasons, they will not be used in amounts higher than about 2 pounds per ton of dry pulp, although in some cases more drainage/retention aid may be used without adverse effect, for instance, 5 or even 10 pounds per ton of dry pulp.

The instant polymers may be applied in any convenient form, e.g., as aqueous solutions, or emulsions, and they may be prepared by emulsion and solution polymerization techniques which are well known to those skilled in the art.

For example, an aqueous phase containing acrylamide and AETAC monomers may be prepared. These monomers are water-soluble. A water-in-oil emulsion of this aqueous phase may then be prepared by dispersing the aqueous phase into a hydrophobic liquid, using a suitable emulsifying surfactant. Inert hydrophobic liquids which can be used as the hydrophobic phase include, but are not limited to, mineral oils, kerosene, naphthas, petroleums and blends of aromatic and aliphatic hydrocarbons. Polymerization is then initiated using a free radical initiator, such as tertiary butyl hydroperoxide. Polymerization may then proceed either adiabatically or isothermally. See, for example, USSN 757,966, which is incorporated by reference into this document, and which describes a method for manufacturing stable water-in-oil emulsions of water soluble polymers using an oil soluble alkanolamide-based surfactant system. Also, see U.S. Patent 1,147,681, which discloses a method for preparing acrylamide-type emulsion polymers.

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## EXAMPLES

The following examples further demonstrate the instant invention. They are not intended to limit the scope of the instant invention in any way.

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### Example 1 - Preparation of Emulsion Polymer

Emulsion polymers of acrylamide and acryloyloxyethyl trimethyl ammonium chloride (AETAC) were prepared by the conventional emulsion polymerization technique described below:

An aqueous phase comprising: a) acrylamide and AETAC in the desired weight ratio; b) water; and c) sodium ethylenediamine tetracetic acid was mixed. The pH of this phase was adjusted to 3.0.

Separately, an oil-soluble alkanolamide-based surfactant system was added to the oil phase. Any conventional emulsifying surfactants can be used. The aqueous phase was then dispersed into the oil/surfactant phase, and polymerization was initiated using a tertiary butyl hydroperoxide/sodium metabisulfate catalyst system under nitrogen. Polymerization continued either adiabatically or isothermally to completion.

Reduced viscosities of the resulting polymers were measured at 0.05 g/l using a 1.0 N sodium chloride solution at 30°C.

For 90/10 acrylamide/AETAC polymers, the aqueous phase comprised:

748.99 g of 45.6% active acrylamide;  
50.60 g of 75% active AETAC;  
262.26 g of water; and  
1.50 g of Na<sub>4</sub> EDTA chelant.

For 97/3 acrylamide/AETAC polymers, the aqueous phase comprised:

828.54 g of 45.6% active acrylamide;  
15.17 g of 75% active AETAC;  
238.14 g of water; and

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1.5 g of Na<sub>4</sub> EDTA chelant.

The oil phase comprised 349.00 g of Kensol 61, a branch-chain hydrocarbon commercially available from Witco Chemical Company, in both the 90/10 and 97/3 cases.

The alkanolamide-based emulsifying surfactant system was added to the oil phase in sufficient amount to uniformly disperse the aqueous phase into the oil phase. All polymers used in Examples 2-20 were prepared in accordance with these instructions.

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## EXAMPLES 2-20

The emulsion polymers of Example 1 were added to various newsprint furnishes to evaluate their effectiveness as retention aids. The furnishes studied are described in Table I, below.

TABLE I  
NEWSPRINT FURNISH

Variable	A	B	C	D	E
Furnish Type	GWD/KFT <sup>1</sup>	CWD/Sulfite <sup>2</sup>	CWD/SBK/TMP/Talc <sup>3</sup>	TMP/Deink Stock	GWD/SBK/TMP
pH	4.4	4.7	4.6	3.5	4.7
Z Consistency (g dry pulp/100 ml)	0.79	0.66	0.99	1.03	1.06
Z Fines	53	55	67	63	67
Z Ash	-	-	3	7	1

GWD is groundwood pulp.

KFT is Kraft pulp.

Sulfite is Kraft sulfite pulp.

SBK is semi-bleached Kraft pulp.

TMP is thermal-mechanical pulp.

To measure retention efficacy, first pass fines retention values were determined using standard Britt Jar Tests. The Britt Jar Tests were run by establishing a mixing speed which gave approximately the same % retention as was obtained on the corresponding paper machine (using untreated headbox stock). The following procedures were followed:

- 5 1. The Britt Jar mixing speed was set at the established rate.
  2. 500 mls of untreated headbox stock were poured into the jar and mixed for 30 seconds.
  3. The pinch clamp was opened and 100 mls of filtrate was collected in a graduated cylinder.
  4. The contents of Step 3 were filtered through pre-weighed filter paper, and then collected fines were completely dried. % fines retention was then calculated, which served as the untreated (comparison  
10 example) % retention value for each furnish.
  5. Retention aids were evaluated by adding the desired amount of polymer to the furnish being tested and mixing. Step 4 was then repeated.
- The results are reported in Table II.

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TABLE II

Example Number	Furnish	Polymer wt/wt AM/AETAC	Polymerization Mode	Reduced Viscosity	Retention Aid Dosage (lbs. active/ ton dry pulp)	% First Pass Fines Retention
2*	C	None	-	-	-	20.8
3	C	90/10	Isothermal Emulsion	20.6	1.0	40.0
4	C	90/10	Isothermal Emulsion	20.4	1.0	40.5
5	C	90/10	Isothermal Emulsion	18.4	1.0	37.0
6	C	90/10	Isothermal Emulsion	16.7	1.0	30.0
7	C	90/10	Isothermal Emulsion	15.4	1.0	36.0
8	C	90/10	Adiabatic Emulsion	14.4	1.0	32.5
9	C	85/15	Adiabatic Emulsion	18.5	1.0	41.0
10*	E	None	-	-	-	17.8
11	E	90/10	Isothermal Emulsion	20.6	1.0	36.5
12	E	90/10	Isothermal Emulsion	27.1	1.0	39.0

12 1 18

12 1 18



TABLE II (continued)

Example Number	Furnish	Polymer wt/wt AM/AETAC	Polymerization Mode	Reduced Viscosity	Retention Aid Dosage (lbs. active/ton dry pulp)	% First Pass Fines Retention
13	E	90/10	Isothermal Emulsion	27.2	1.0	42.4
14	E	90/10	Isothermal Emulsion	28.4	1.0	43.2
15*	A	None	-	-	-	36.0
16	A	97/3	Isothermal Emulsion	22.0	1.0	52.0
17*	B	None	-	-	-	33.0
18	B	97/3	Isothermal Emulsion	22.0	1.0	51.0
19*	D	None	-	-	-	25.0
20	D	97/3	Isothermal Emulsion	26	1.0	49.0

\*Comparison Example

Examples 21 - 24

The effectiveness of the instant polymers as drainage aids was evaluated using a standard Schopper-Riegler Freeness Tester. Standard laboratory procedures were employed.

An unbleached Kraft/waste furnish corrugating media served as the test furnish. This furnish had a % consistency of 1.28, a pH of 7.5 and a stock temperature of 120°F. The polymer dosage was 4.0 lbs/ton of dry pulp.

Results are shown in Table III.

TABLE III

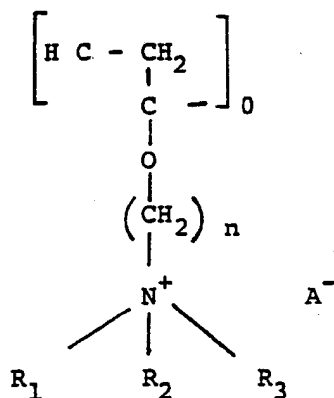
**DRAINAGE PERFORMANCE OF AA/AETAC POLYMERS  
IN A CORRUGATED FURNISH**

<u>Example Number</u>	<u>AM/AETAC Drainage Aid (Wt/Wt)</u>	<u>Reduced Viscosity (dl/g)</u>	<u>% Increase in Freeness over No Aid</u>
21	97/3	26.2	15.5
22	95/5	24.5	23.0
23	90/10	28.4	34.0
24	85/15	27.2	20.5

**Claims**

1. A method for improving drainage and/or retention of inorganic fillers, pigments and/or cellulosic fines in paper or paper-like product comprising adding to a furnish used to prepare said paper or paper-like product an effective amount of a polymer comprising:

- (a) 75 to 99.9%, by weight, acrylamide or methacrylamide, and  
(b) 25 to 0.1%, by weight, a monomer of the following structure:



wherein n is 2 or 3, wherein R<sub>1</sub>, R<sub>2</sub>, R<sub>3</sub>, which may be the same or different, are C<sub>1</sub>-C<sub>8</sub> alkyl; and wherein A is -Cl, Br, or CH<sub>3</sub>SO<sub>3</sub>; wherein said polymer has a reduced viscosity of from about 18 to about 30



dl/g at 0.05 weight percent using 1 N NaCl at 30°C.

2. The method of Claim 1, wherein said effective amount is from about 0.1 to about 10 pounds polymer per ton of dry pulp.

3. The method of Claim 1, wherein (a) is acrylamide and wherein (b) is acryloyloxyethyl trimethyl ammonium chloride.

4. The method of Claim 3, wherein (2) is 85 to 99%, by weight, of said polymer and (b) is 1 to 15%, by weight of said polymer.

5. The method of Claim 6, wherein said paper or paper-like product is newsprint.

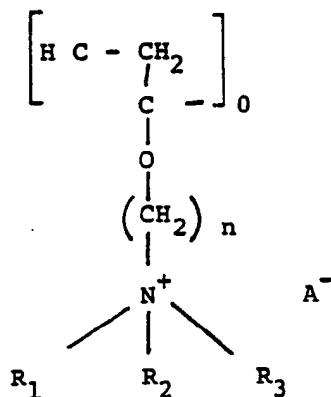
6. A composition comprising:

(a) a paper or paper-like furnish; and

(b) an effective amount of a polymer comprising:

(i) 75 to 99.9%, by weight, acrylamide or methacrylamide, and

(ii) 25 to 0.1%, by weight, a monomer of the following structure:



wherein n is 2 or 3; wherein R<sub>1</sub>, R<sub>2</sub> and R<sub>3</sub>, which may be the same or different, are C<sub>1</sub>-C<sub>8</sub> alkyl; wherein A is Cl, Br or CH<sub>3</sub>SO<sub>4</sub>; and wherein said polymer has a reduced viscosity of from about 18 to about 30 dl/g at 0.05 weight percent using 1 N BaCl<sub>2</sub> at 39°C.

7. The composition of Claim 14, wherein said effective amount is from about 0.1 to about 10 pounds polymer per ton of dry pulp.

8. The composition of Claim 14, wherein said paper or paper-like furnish is a newsprint furnish.

9. The composition of Claim 16, wherein (i) is acrylamide and wherein (ii) is acryloyloxyethyl trimethyl ammonium chloride.

10. The composition of Claim 17, wherein (i) 85 to 99%, by weight, of said polymer and wherein (ii) is 1 to 15%, by weight, of said polymer.

⑫

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10.08.88 Bulletin 88/32

⑱ Designated Contracting States:  
BE CH DE FR GB LI SE

⑲ Date of deferred publication of search report:  
17.08.88 Bulletin 88/33

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## EUROPEAN SEARCH REPORT

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Application Number

EP 88 30 0477

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl. 4)
A	US-A-3 239 496 (M.J. JURSIK)		
A	US-A-2 838 397 (I.J. GRUNTFEST et al.)		
A	GB-A-1 195 737 (NALCO CHEMICAL CO.)		
			TECHNICAL FIELDS SEARCHED (Int. Cl. 4)
The present search report has been drawn up for all claims			
Place of search THE HAGUE		Date of completion of the search 04-05-1988	Examiner NESTBY K.
CATEGORY OF CITED DOCUMENTS		T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons ----- & : member of the same patent family, corresponding document	
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Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl. 4)
X	US-A-4 581 402 (W. DUNK et al.) * Claims 1,2,5-7; examples 3,6 * ---	1,2,4,6 ,7,10	D 21 H 3/38
X	US-A-4 461 866 (M.J. YANUTOLA) * Examples 1,3,8 * ---	1,2,6,7	
X	US-A-3 901 857 (G. SACKMANN et al.) * Whole document * ---	1-4,6,7 ,9,10	
X	TAPPI JOURNAL, vol. 66, no. 6, June 1983, pages 83-85, Atlanta, Georgia, US; T. LINDSTRÖM et al.: "Effects of pH and electrolyte concentration on the adsorption of cationic polyacrylamides on cellulose" * Whole article * ---	1,2,6,7	
X	ABSTRACT BULLETIN OF THE INSTITUTE OF PAPER CHEMISTRY, vol. 51, no. 10, April 1981, page 1111, abstract no. 10392, Appleton, Wisconsin, US; T. MIWA et al.: "Improvement of filler efficiency in manufacturing paper" * Whole abstract * ---	1-3,6,7 ,9,10	TECHNICAL FIELDS SEARCHED (Int. Cl.4)
A	ABSTRACT BULLETIN OF THE INSTITUTE OF PAPER CHEMISTRY, vol. 48, no. 5, November 1977, page 527, abstract no. 4846; K. SUZUKI et al.: "Preparation of cationic polymers and their applications. (6). Effect of partially aminated polyacrylamides on paper strength improvement for various pulps (especially for newsprint paper pulp)", & JAPAN TAPPI 31, no. 5: 270-274 (May 1977) * Whole abstract * ---	5,8	D 21 H
The present search report has been drawn up for all claims			
Place of search THE HAGUE		Date of completion of the search 04-05-1988	Examiner NESTBY K.
<b>CATEGORY OF CITED DOCUMENTS</b> X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document			

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